CLAIMS

This listing of the claims will replace all prior versions, and listings of claims in the Application.

- (currently amended) One-part self-etching, self-priming dental adhesive composition having a pH of at most 2, which comprises-comprising:
 - (a) a polymerizable acidic phosphoric acid ester monomer of the following formula
 (A):

$$Y = \begin{pmatrix} 0 & 0 \\ HO & O-Y \end{pmatrix}_{b}$$
 (A)

wherein the moieties Y independent from each other represent a hydrogen atom or a moiety of the following formula (Y)

$$\left(z_{i}\right)$$
 O C (Y)

wherein

 Z_1 is $COOR^{10}$, $COSR^{20}$, $CON(R^{10})_2$, $CONR^{10}R^{20}$, or $CONHR^{10}$, wherein R^{10} and R^{20} independently represent

- a hydrogen atom,
- a C_{1-18} alkyl group optionally substituted by a C_{3-8} cycloalkyl group,
- an optionally substituted C₃₋₈ cycloalkyl group,
- an optionally substituted C4-18 aryl or heteroaryl group,
- an optionally substituted C_{5-18} alkylaryl or alkylheteroaryl group, or
- an optionally substituted C_{7-30} aralkyl group,

whereby two R_1 residues may form together with the adjacent nitrogen atom to which they are bound a 5- to 7-membered heterocyclic ring which may contain further

nitrogen atoms or an oxygen atoms, and whereby the optionally substituted groups may be

substituted by 1 to 5 C₁₋₅ alkyl groups; group(s);

L represents an (a+b)-valent organic residue, [[(]]whereby b is 1 when Y in formula (A) is within the round brackets[[is]]_L containing 2 to 45 carbon atoms and optionally heteroatoms, euch-ae oxygen, nitrogen and sulfur atoms, the carbon atoms including a+b carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said a+b carbon atoms linking a phosphate or 2-(oxa-ethyl)acryl derivative group; a is an integer of from 1 to 10-preferably-1-to-6; b is an integer of from 1 to 10-preferably-1-to-6;

provided that at least one Y is not hydrogen; and

- one or more polymerisable acidic monomers selected from the group consisting of
 - (b1) polymerisable acidic monomers of the following formula (B):

$$\left(\begin{array}{ccc}
R1 & & & & \\
R2 & & & & \\
R2 & & & & \\
\end{array}\right) L_1 \left(\begin{array}{c} C & & & \\ P & OH \\ OH \end{array}\right)_d$$
(B)

wherein

R₁ and R₂ independently represent

a hydrogen atom.

an optionally substituted C1-18 alkyl group,

an optionally substituted Cats cycloalkyl group.

an optionally substituted C₅₋₁₈ arvl or heteroarvl group.

an optionally substituted C₅₋₁₈ alkylaryl or alkylheteroaryl group.

an optionally substituted C7-30 aralkyl group,

whereby the optionally substituted groups may be substituted by 1 to 5 $C_{1.5}$ alkyl groups; group(e);

L, represents a (c + d) valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur, the carbon atoms including c + d carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said c+d carbon atoms linking a phosphonate or optionally substituted acrylamido group;

and

c and d independently represent integers of from 1 to 10;

(b2) polymerisable acidic monomers of the following formula (C):

wherein

Z₂ independently has the same meaning as defined for Z₁;

 L_2 represents an (e+f) valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including e+f carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said e+f carbon atoms linking a sulphonate or optionally substituted 2-(oxa-ethyl)acryl derivative group; and e+f and f independently represent an integer of from 1 to 10;

(b3) acidic monomers of the following formula (D):

$$\begin{pmatrix}
R3 & N & L_3 & Q & OH \\
R4 & C & Q & OH
\end{pmatrix}$$
(D)

wherein

R₃ and R₄ independently represent

a hydrogen atom,

an optionally substituted C1718 alkyl group,

an optionally substituted C3-18 cycloalkyl group,

an optionally substituted C5"18 aryl or heteroaryl group,

an optionally substituted C₅₋₁₈ alkylaryl or alkylheteroaryl group,

an optionally substituted C7-30 aralkyl group,

whereby the optionally substituted groups may be substituted by 1 to 5 C₁
• alkyl groups: group(s)

 L_3 represents a (g+h) valent valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including g+h carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and

aromatic carbon atoms, each of said g+h carbon atoms linking a sulphonate or optionally substituted acrylamido group; and g and h independently represent integers ef-from 1 to 10;

- (c) a polymerizable N-substituted alkylacrylic or acrylic acid amide monomer;
- (d) optionally an organic and/or inorganic acid;
- (e) an organic water soluble solvent and/or water; and
- (f) <u>a</u> polymerization initiator[[,]];
- (g) an inhibitor; and
- (h) a stabilizer.
- (original) The one-part self-etching, self-priming dental adhesive composition of claim 1
 wherein L₁, L₂, and L₃ independently represent
 an optionally substituted C₁₋₁₈ alkylene group which may contain from 1 to 9 oxygen

atoms in the chain,
an optionally substituted C₂₇₁₈ cycloalkylene group.

an optionally substituted C₅₋₁₈ arviene or heteroarviene group.

an optionally substituted C₅₋₁₈ alkylaryl or alkylheteroarylene group,

an optionally substituted C₇₋₃₀ aralkylene group.

- (currently amended) The one-part self-etching, self-priming dental adhesive composition
 of claim 1 or 2 which is hydrolysis stable for at least one week at a storage temperature
 of 50 °C, whereby after such storage the bond strength of an adhesive prepared from
 such an adhesive composition to enamel and/or dentin is at least 10 MPa, preferably 15
 MPa.
- (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any one of the preceding claims, wherein components (a) and (b) are contained in a ratio of from 1:100 to 100: 1.
- 5. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any-one-of the preceding claims, wherein said organic acid of component (d) is selected from the group <u>consisting</u> of mono- or polycarboxylic acids, <u>such-as</u> methacrylic acid, acrylic acid, fumaric acid, maleic acid, citric acid, itaconic acid,

and formic acid, and wherein the inorganic acid of component (d) is selected from the group consisting of sulfonic acid, phosphoric acid, sulfuric acid and hydrofluoric acid.

- (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any one of the preceding-claims, wherein said organic water soluble solvent of component (e) is selected from the group <u>consisting</u> of alcohols, and ketones, <u>euch as</u> ethanol, propanol, butanol, acetone, <u>and</u> methyl ethyl ketone.
- (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any-one-of the preceding-claims, wherein said acidic polymerizable monomer of component (a) is characterized by one of the following formulas:

wherein

Z or Z₁ is $COOR^{10}$, $COSR^{20}$, $CON(R^{10})_2$, $CONR^{10}R^{20}$, or $CONHR^{10}$, wherein R^{10} and R^{20} independently represent a hydrogen atom.

a C_{1-18} alkyl group optionally substituted by a C_{3-8} cycloalkyl group, an optionally substituted C_{3-8} cycloalkyl group, an optionally substituted C_{4-18} aryl or heteroaryl group, an optionally substituted C_{5-18} alkylaryl or alkylheteroaryl group, or an optionally substituted C_{7-30} aralkyl group, whereby two R_1 residues may form together with the adjacent nitrogen atom to which they are bound a 5- to 7-membered heterocyclic ring which may contain further nitrogen atoms or an oxygen atoms,

and whereby the optionally substituted groups may be substituted by 1 to 5 C_{1-5} alkyl groups; group(s);

L represents an (a+b)-valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including a + b carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said a+b carbon atoms linking a phosphate or 2-(oxa-ethyl)acryl derivative group; a is an integer ef from 1 to 10, preferably 1 to 5; b is an integer ef from 1 to 10, preferably 1 to 5

wherein Z is as defined in claim 1 and

- (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any one of the preceding claims; wherein said acidic polymerizable monomer of component (b) is a polymerisable acidic monomers of formula (C).
- (currently amended) Hydrolysis-stable The one-part self-etching, self-priming dental adhesive composition of claim 8, wherein said acidic polymerizable monomer is characterized by one of the following formulas:

10. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any-one-of the preceding-claims, wherein the polymerizable Nsubstituted alkylacrylic or acrylic acid amide monomer of component (c) is characterized by one of the following formulas:

wherein

R₅ and R₆ independently represent

a hydrogen atom or a substituted

a C₁ to C₁₈ alkyl group,

an optionally substituted C3-18 cycloalkyl group,

an optionally substituted C5-18 aryl or heteroaryl group,

an optionally substituted C₅₋₁₈ alkylaryl or alkylheteroaryl group,

an optionally substituted C7-30 aralkyl group,

R₇ represents a

a divalent substituted or unsubstituted organic residue having from 1 to 45 carbon atoms, whereby said organic residue may contain from 1 to 14 oxygen and/or nitrogen atoms and is selected from a C₁ to C₁₆ alkylene group wherein from 1 to 6 -CH₂-groups may be replaced by a -N-(C=O)-CR₉=CH₂ group wherein R₉ is a

hydrogen atom or a C $_1$ to C $_{18}$ alkyl group, a divalent substituted or unsubstituted C $_3$ to C $_{18}$ cycloalkyl or cycloalkylene group, a divalent substituted or unsubstituted C $_4$ to C $_{18}$ aryl or heteroaryl group, a divalent substituted or unsubstituted C $_5$ to C $_{18}$ alkylaryl or alkylheteroaryl group, a divalent substituted or unsubstituted C $_7$ to C $_{30}$ aralkyl group, and a divalent substituted or unsubstituted C $_2$ to C $_{45}$ mono-, di- or polyether group having from 1 to 14 oxygen atoms,

R₈ represents

a saturated di- or multivalent substituted or unsubstituted cyclic C₁₈ hydrocarbon group, a saturated di- or multivalent substituted or unsubstituted cyclic C₃ to C₁₈ hydrocarbon group, a di- or multivalent substituted or unsubstituted C₄ to C₁₈ anyl or heteroaryl group, a di- or multivalent substituted or unsubstituted C₅ to C₁₈ alkylaryl or alkylheteroaryl group, a di- or multivalent substituted or unsubstituted C₇ to C₃₀ aralkyl group, or a di- or multivalent substituted or unsubstituted C₂ to C₄₅ mono-, di-, or polyether residue having from 1 to 14 oxygen atoms, and n is an integer.

11. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any one of the preceding claims, wherein said polymerizable monomer is a mono-, bis- or poly(meth) acrylamide characterized by one of the following formulas:

- (currently amended) The one-part self-etching, self-priming dental adhesive composition
 according to <u>claim 1, any-one-of the-preceding-claims</u>, which contains said acidic
 polymerizable monomers of components (a) and (b) in an amount ef-from 5 to 90 wt-%.
- (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any one of the preceding-claims, wherein said polymerization initiator is a thermal initiator, a redox-initiator or a photo initiator.
- 14. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 13</u>, any one of the preceding claims, wherein said photo initiator is champhor quinone.
- 15. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any-one-of-the-preceding-claims, wherein said filler is an inorganic filler and/or an organic filler; preferably the filler is a nanofiller.
- (currently amended) The one-part self-etching, self-priming dental adhesive composition
 according to <u>claim 1</u>, any one of the preceding claims, wherein said stabilizer is a radical
 absorbing monomer, such as hydroquinone, hydroquinone monomethylether, 2,6-di-tert.butyl-p-cresol.

 (currently amended) The one-part self-etching, self-priming dental adhesive composition according to claim 1, any one of the preceding claims, wherein L represents

an (a+b)-valent saturated aliphatic C_2 to C_{18} group having at least 2 of said primary aliphatic carbon atoms, and optionally 1 or more of said secondary aliphatic carbon atom(s), whereby said (a+b)-valent group may be substituted by C_1 to C_5 alkyl group(s); or

a C_2 to C_{45} mono-, di-, or polyether which has from 1 to 14 oxygen atoms and is substituted by at least 2 C_1 to C_{10} aliphatic $\underline{\text{groups group($+$)}}$ having said primary and/or secondary aliphatic carbon atoms; whereby said ether may optionally be substituted by C_1 to C_5 alkyl $\underline{\text{groups:group($+$)}}$; or

wherein L represents:

a saturated C_3 to C_8 cyclic, C_7 to C_{15} bi- or polycyclic hydrocarbon group having from 0 to 4, preferably, 0 to 3, more preferably 0 or 1, of said secondary alicyclic carbon atoms: and/or

a C₄ to C₁₈ aryl or heteroaryl group having from 0 to 5, preferably 0 to 3, more preferably 0 or 1, of said aromatic carbon atoms; whereby said saturated hydrocarbon or aryl or heteroaryl group is substituted by

from 0 to 5 C₁ to C₅ alkyl groups; group(s);

from 0 to 4, preferably 1 to 3, more preferably 1 or 2, saturated C_1 to C_{10} aliphatic group(s) having said primary and/or secondary aliphatic carbon atoms, and/or from 0 to 2 divalent residues according to one of the following formulas:

-[O-CH₂CH₂-]_r- wherein f is an integer of from 1 to 10, preferably 1 to 5;

-[-O-CH $_2$ CH $_2$ CH $_2$ -] $_9$ - wherein g is an integer from effrem 1 to 10, preferably 1 to 5; -[O-R $_{12}$] $_n$ - wherein R $_{12}$ is -CH(CH $_3$)-CH $_2$ - or -CH $_2$ -CH(CH $_3$)- and h is an integer of from 1 to 10, preferably 1 to 5;

-[-O-R₁₄]-[O-R₁₅]_r or -[O-R₁₅]_x-[O-R₁₄]_r wherein R₁₄ is -CH₂CH_{2*}-, R₁₅ is -CH(CH₃)- CH_{2*}- or -CH₂-CH(CH₃)-, i, j, k, and I are integers whereby

 $2i + 3j \le 15$ and $2k + 3l \le 15$,

-[O-CH2CH2CH2CH2-]r- wherein r is an integer of 1 or 2;

wherein said divalent residues have one of said primary aliphatic carbon atoms; and

whereby 2 groups selected from said saturated hydrocarbon, aryl, and heteroaryl groups may optionally be linked by a single bond, an alkylene group, or -O-.

18. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, <u>any-one-of-the-preceding-claims</u>, wherein L represents an (a+b)-valent saturated C₃ to C₆ cyclic or C₇ to C₁₅ bi- or tricyclic hydrocarbon group having at least 2 of said secondary alicyclic carbon atoms; an (a+b)-valent saturated C₄ to C₁₉ aryl or heteroaryl group having from 2 to 6 of said aromatic carbon atoms; an (a+b)-valent C₆ to C₁₈ alkylaryl or alkyl heteroaryl group having at least one of said aromatic carbon atoms, at least one of said secondary aliphatic carbon atoms, and optionally one of said primary aliphatic carbon atoms at the terminal end of the alkyl

an (a+b)-valent C_8 to C_{30} aralkyl group having at least one of said primary aliphatic carbon atoms and at least one of said secondary aliphatic carbon atoms.

mojety of said alkylaryl or alkylheteroaryl group; or

19. (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any one of the preceding claims, wherein L represents is a divalent residue according to one of the following formulas:

$$\begin{split} & -[CH_2CH_2-O_-]_m-CH_2CH_2- \text{ wherein } m \text{ is an integer ef from 1 to 14,} \\ & -[CH_2CH_2CH_2-O_-]_p-CH_2CH_2 CH_2- \text{ wherein p is an integer ef from 1 to 14,} \\ & -[R_{12^*}O]_{q^*}R_{13^*} \text{ wherein } R_{12} \text{ and } R_{13} \text{ may be } -CH(CH_3)-CH_2- \text{ or } \\ & -CH_2-CH(CH_3)- \text{ and q is from 1 to 14,} \\ & -[R_{14^*}O]_r-[R_{15^*}O]_5-R_{14^*} \text{ or } -[R_{14^*}O]_r-[R_{15^*}O]_0-R_{15^*} \text{ wherein } R_{14} \text{ is } \\ & -CH_2CH_2-R_{15} \text{ is } -CH(CH_3)-CH_2- \text{ or } -CH_2-CH(CH_3)-, r, s, t, \text{ and u are integers } \\ & \text{whereby } 2r+3s \leq 43 \text{ and } 2t+3u \leq 42, \\ & -[CH_2CH_2CH_2CH_2CH_2-O_r-CH_2CH_2CH_2- \text{ wherein r is 1 or 2,} \end{split}$$

$$-CH_{2} - CH_{2} -$$

wherein R_{18} and R_{17} are H or -CH₃ and x and y may independently be integers of from 0 to 10, preferably 0 to 5.

- (currently amended) The one-part self-etching, self-priming dental adhesive composition
 according to <u>claim 1</u>, any one of the preceding-claims, wherein said (a+b) carbon atoms
 are primary aliphatic carbon atoms.
- (currently amended) The one-part self-etching, self-priming dental adhesive composition
 according to <u>claim 1</u>, any one of the preceding claims, wherein the polymerizable acidic
 phosphoric acid ester monomer is of the following formula (A-1):

wherein

or

 Z_1 is $COOR^{10}$, $COSR^{20}$, $CON(R^{10})_2$, $CONR^{10}R^{20}$, or $CONHR^{10}$, wherein

R¹⁰ and R²⁰ independently represent

a hydrogen atom,

a $C_{1^{-1}8}$ alkyl group optionally substituted by a $C_{3^{-8}}$ cycloalkyl group.

an optionally substituted C3-8 cycloalkyl group.

an optionally substituted C4-18 aryl or heteroaryl group,

an optionally substituted $C_{5^{\sim}18}$ alkylaryl or alkylheteroaryl group, or

an optionally substituted C7-30 aralkyl group,

whereby two R_1 residues may form together with the adjacent nitrogen atom to which they are bound a 5- to 7-membered heterocyclic ring which may contain further nitrogen atoms or an oxygen atoms.

and whereby the optionally substituted groups may be substituted by 1 to 5 C₁₋₅ alkyl groups: eroup(s):

L represents an (a+b)-valent organic residue containing 2 to 45 carbon atoms and optionally heteroatoms, euch as oxygen, nitrogen and sulfur atoms, the carbon atoms including a+b carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said a+b carbon atoms linking a phosphate or 2-(oxa-ethyl)acryl derivative group;

- a is an integer of from 1 to 10, preferably 1 to 5;
- b is an integer of from 1 to 10, preferably 1 to 5.
- (currently amended) The one-part self-etching, self-priming dental adhesive composition according to <u>claim 1</u>, any one-of-claims 1 to 20 wherein none of the moieties Y is a hydrogen atom.
- (currently amended) A polymerizable acidic phosphoric acid ester monomer of the following formula (A)

$$Y = \begin{pmatrix} 0 & 0 \\ 0 & 0 - Y \end{pmatrix}_{b}$$
 (A)

wherein

the moieties Y independent from each other represent a moiety of the following formula (Y)

$$\left(z_{i}\right)$$
 O L (Y)

wherein

 Z_1 is $COOR^{10}$, $COSR^{20}$, $CON(R^{10})_2$, $CONR^{10}R^{20}$, or $CONHR^{10}$, wherein R^{10} and R^{20} independently represent

a hydrogen atom,

a $C_{1^{-1}8}$ alkyl group optionally substituted by a $C_{3^{-8}}$ cycloalkyl group,

an optionally substituted $C_{3^{-8}}$ cycloalkyl group, an optionally substituted $C_{4^{-18}}$ aryl or heteroaryl group, an optionally substituted $C_{5^{-18}}$ alkylaryl or alkylheteroaryl group. or

an optionally substituted $C_{7^{-90}}$ aralkyl group, whereby two R_1 residues may form together with the adjacent nitrogen atom to which they are bound a 5- to 7-membered heterocyclic ring which may contain further nitrogen atoms or an oxygen atoms,

and whereby the optionally substituted groups may be substituted by 1 to 5 C₁₋₅ alkyl groups; group(s);

L represents an (a+b)-valent organic residue, [[(]]whereby b is 1 when Y in formula (A) is within the round brackets[[is]]]. L containing 2 to 45 carbon atoms and optionally heteroatoms, such as oxygen, nitrogen and sulfur atoms, the carbon atoms including a+b carbon atoms selected from primary and secondary aliphatic carbon atoms, secondary alicyclic carbon atoms, and aromatic carbon atoms, each of said a+b carbon atoms linking a phosphate or 2-(oxa-ethyl)acryl derivative group;

a is an integer of from 1 to 10, preferably 1 to 5;

b is an integer of from 1 to 10, preferably 1 to 5, more preferably 1.